

PATENT COOPERATION TREATY

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From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

PCT

To:

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NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Rule 71.1)

Date of mailing
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03.05.2004

Applicant's or agent's file reference
2002B044A

IMPORTANT NOTIFICATION

International application No.
PCT/US 03/09733

International filing date (day/month/year)
28.03.2003

Priority date (day/month/year)
29.03.2002

Applicant
EXXONMOBIL CHEMICAL PATENTS INC.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.
4. **REMINDER**

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

The applicant's attention is drawn to Article 33(5), which provides that the criteria of novelty, inventive step and industrial applicability described in Article 33(2) to (4) merely serve the purposes of international preliminary examination and that "any Contracting State may apply additional or different criteria for the purposes of deciding whether, in that State, the claimed inventions is patentable or not" (see also Article 27(5)). Such additional criteria may relate, for example, to exemptions from patentability, requirements for enabling disclosure, clarity and support for the claims.

Name and mailing address of the international preliminary examining authority:



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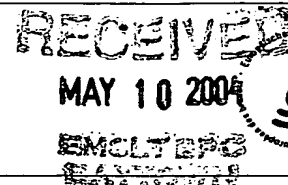
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INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference 2002B044A		FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/PEA416)
International application No. PCT/US 03/09733	International filing date (<i>day/month/year</i>) 28.03.2003	Priority date (<i>day/month/year</i>) 29.03.2002
International Patent Classification (IPC) or both national classification and IPC C07C2/12, C07C2/12		
Applicant EXXONMOBIL CHEMICAL PATENTS INC.		
<p>1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of 6 sheets, including this cover sheet.</p> <p><input checked="" type="checkbox"/> This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).</p> <p>These annexes consist of a total of 9 sheets.</p>		
<p>3. This report contains indications relating to the following items:</p> <p>I <input checked="" type="checkbox"/> Basis of the opinion</p> <p>II <input type="checkbox"/> Priority</p> <p>III <input type="checkbox"/> Non-establishment of opinion with regard to novelty, inventive step and industrial applicability</p> <p>IV <input type="checkbox"/> Lack of unity of invention</p> <p>V <input checked="" type="checkbox"/> Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement</p> <p>VI <input type="checkbox"/> Certain documents cited</p> <p>VII <input type="checkbox"/> Certain defects in the international application</p> <p>VIII <input type="checkbox"/> Certain observations on the international application</p>		
Date of submission of the demand 09.09.2003		Date of completion of this report 03.05.2004
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465		Authorized Officer Seelmann, M Telephone No. +49 89 2399-8335 

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. **PCT/US 03/09733**

I. Basis of the report

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

Description, Pages

1-10 as originally filed

Claims, Numbers

1-21

filed with telefax on **16.02.2004**
~~15.01.2004~~

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

**INTERNATIONAL PRELIMINARY
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**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability;
citations and explanations supporting such statement**

1. Statement

Novelty (N)	Yes: Claims	1-21
	No: Claims	
Inventive step (IS)	Yes: Claims	
	No: Claims	1-21
Industrial applicability (IA)	Yes: Claims	1-21
	No: Claims	

2. Citations and explanations

see separate sheet

The present application relates to the production of a C12+ fraction of olefinic hydrocarbons by oligomerizing a lower olefinic hydrocarbon feedstock in the presence of a surface-deactivated ZSM-23 catalyst.

D1 US 5 026 933

D2 US 4 855 527

D3 US 5 284 989

D4 US 3 440 291

D5 FR 2 527 201

D6 G. Marceglia *et al.*, Chem. Eco. Engin. Rev., 14(6), 35-40 (1982)

Item V

Reasoned statement under Artikel 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

V.1 Novelty

Olefin oligomerization with the surface modified zeolite catalyst ZSM-23 is already known from the prior art (**D1-D3**). These oligomers obtained are also further separated into fractions by conventional distillation separation, *i.e.* C12+, C15+, C16+ or C18+. Branching is also encountered, which outlined the presence of iso-olefins (even to a minimum extent) in the starting olefin feedstock. The produced olefin oligomers are also known to be used in a process for the production of long chain alcohol (**D4**, table 2, line 9), as alkylating agents in the selective alkylation of an aromatic compound or in the production of alkylphenylsulfonates using the previously produced alkylbenzenes as intermediates (**D1-D3**). Accordingly even though the prior art documents **D1-D3** are silent about the content of the iso-olefin in the C₂-C₆ olefin feedstock used as starting material, due to the occurrence of branching, novelty could only be recognized based on a selection invention in view **D1-D3**, *i.e.* the present application specifies a content of about 0.5 wt% to about 25 wt% of an iso-olefin in the initial olefin feedstock !

V.2 Inventive step

The present application (claims 1-21) does not involve an inventive step for the following reasons:

The technical problem is to provide an improved process to produce slightly branched, high

molecular weight olefinic hydrocarbons. The proposed solution is the use of a surface-deactivated ZSM-23 catalyst and the separation of the C12+ fraction, which is characterized by a quaternary carbon content of less than 0.5 atom%.

- 2₁. No examples are present in the present application to illustrate that such a technical problem has been solved and that the content in iso-olefin in the starting feedstock is an essential feature.
- 2₂. It is questionable that such a process would solve the technical problem posed over the entire scope of protection claimed, since it is specified in claim 1 that the result of the C12+ fraction, which is characterized by a quaternary carbon content of less than 0.5 atom%, is obtained under any possible oligomerization conditions with ZSM-23 as catalyst. This claim does not specify which specific experimental conditions, of temperature or pressure for instance, are necessary to achieve such a result.
- 2₃. A man skilled in the art already knows that a feedstock comprising C₂-C₈ n-olefins does also possess dienes, iso-olefins and so-on (D6). The olefins used in D1-D3 are nowhere to be said to be pure and leads also to branching under oligomerization conditions with ZSM-23, so that the presence of iso-olefins even though not explicitly disclosed, is implicitly to be recognized. So using the same starting material under the same experimental conditions, one would expect that the same result would be obtained. The technical problem is accordingly already solved in D1-D3.
- 2₄. Novelty was only acknowledged based on a selection invention over D1-D3. An inventive step can only be acknowledged if a surprising effect or property is shown for such a process. Without experimental proof showing the relevance of about 0.5 wt% to about 25 wt% of an iso-olefin in the initial feedstock for the recovery of oligomers with less than 0.5 atom % of quaternary carbons, no inventive step is recognized.

Further comments:

1. Claim 1 does not meet the requirements of Article 6 PCT in that the matter for which protection is sought is defined in terms of the result to be achieved, i.e. the content of the quaternary carbon atoms in the C12+ fraction of the oligomerized olefin, which merely amounts to a statement of the underlying problem. This results depends actually on the oligomerization conditions used, which should have been specified in this claim.

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/US 03/09733

2. The use of the word "about" especially in connection with numerical ranges is generally regarded as rendering the determination of the exact scope of the range difficult (claim 1: wt% of the iso-olefin). When used in a claim, this results in a lack of clarity, contrary to Article 6 PCT.

use of an olefin feedstock that contains significant quantities of iso-olefins, such as isobutylene, without producing deleterious quantities of quaternary carbon atoms in the C₁₂+ fraction.

[0019] The olefinic hydrocarbon feedstock used in the process of the invention comprises one or more C₂ to C₆ n-olefins, such as propene and/or n-butene. In addition, the feedstock contains about 0.1% to about 25%, such as about 0.5% to 5%, of an iso-olefin by weight of the total feedstock. Typically, the iso-olefin will be iso-butylene and/or iso-amylene. One preferred olefinic feedstock for use in the process of the invention is the unreacted effluent stream from an MTBE production unit, which stream typically contains n-butene together with iso-butylene in amounts up to 5 wt%. A practical feedstock, such as an MTBE effluent, may also contain dimethyl ether and sulfur impurities. If present, the dimethyl ether content is preferably less than 100 ppm and the sulfur content is preferably less than 10 ppm.

[0020] The olefinic hydrocarbon feedstock can also contain low molecular weight, typically C₄-C₆, saturated hydrocarbons, typically in amounts between about 5% and about 70% by weight of the overall feedstock.

[0021] The oligomerization catalyst used in the process of the invention comprises ZSM-23 which has been surface deactivated, typically by treatment with a sterically hindered nitrogenous base, such as a trialkyl pyridine compound, and preferably with 2,4,6-collidine (2,4,6-trimethyl pyridine, gamma-collidine).

The surface deactivating compound should have a minimum cross-sectional diameter greater than the effective pore size of the zeolite to be treated; i.e., greater than 5 Angstroms. ZSM-23 and its characteristic X-ray diffraction pattern are described in detail in U.S. Patent No. 4,076,842. Preferably, the ZSM-23 employed in the catalyst has an alpha value of about 25 and a crystal size of less than 0.1 micron and is conveniently composited with a binder, such as alumina.

[0022] Suitable oligomerization conditions include a temperature of about 160°C to about 250°C, such as about 190°C to about 230°C, for example about 210°C to about 220°C; a pressure in the range of about 500 psig (3447 kPa (gauge)) to about 1500 psig (10342 kPa (gauge)), such as in the range of about

750 psig (5171 kPa (gauge)) to about 1250 psig (8618 kPa (gauge)), and a feed weight hour space velocity (WHSV) in the range of about 0.1 hr^{-1} to about 4.0 hr^{-1} , such as in the range of about 0.2 hr^{-1} to about 3.0 hr^{-1} , for example in the range of about 1.75 hr^{-1} to about 2.25 hr^{-1} .

[0023] Where surface deactivation is achieved by treatment with a trialkyl pyridine compound, the feed to the oligomerization process can include additional trialkyl pyridine compound so that the surface properties of the zeolite are maintained during the process. Further details of the oligomerization process can be found in U.S Patent No. 5,026,933.

[0024] The product of the oligomerization process of the invention is an olefinic hydrocarbon mixture which comprises at least 5 wt%, such as at least 20 wt%, for example at least 85 wt% of mono-olefin oligomers of the empirical formula:



wherein n is greater than or equal to 6 and wherein said mono-olefin oligomers comprise at least 20 wt%, and conveniently at least 60 wt%, of olefins having at least 12 carbon atoms and said olefins having at least 12 carbon atoms ($\text{C}_{12}+$ olefins) have an average of from about 0.8 to about 2.0, such as from about 0.8 to about 1.3, C_1 - C_3 alkyl branches per carbon chain. Conveniently, the olefins having at least 12 carbon atoms contain no branches other than methyl and ethyl groups.

[0025] In particular it is found that, despite the presence of iso-olefins in the oligomerization feed, the $\text{C}_{12}+$ olefinic product of the present process contains less than 0.5 atom%, of quaternary carbon atoms. As previously stated, although the reasons for the low quaternary carbon content of the $\text{C}_{12}+$ olefinic product are not fully understood, it is believed that the size of the pores of the ZSM-23 are such that, although iso-butylene can enter the pores to react with, for example, n -butylene, the resultant branched C_8 oligomer is too large to access the pores for further reaction. Thus the iso-olefin reaction products are concentrated, in the case of a C_4 olefin feed, in the C_8 fraction. Because such a fraction inherently has a high octane value, it is advantageous to remove this fraction from the

oligomerization product for use as a gasoline blending component.

[0026] The percentage of quaternary carbon atoms in the $C_{12}+$ olefinic product is conveniently determined by the ^{13}C -NMR technique described in U.S. Patent No. 5,849,960 at column 4, line 23 to column 5, line 3 and in particular the J-Modulated Spin Echo NMR technique (JMSE) using a $1/2J$ delay of 4 ms and incorporating the DEPT-135 NMR correction.

[0027] The lightly branched $C_{12}+$ olefinic hydrocarbon fraction from the oligomerization process of the invention is conveniently used in the production of long chain alcohols for application as, for example, detergents, soaps, surfactants, and freeze point depressants in lubricating oils. Typically this is achieved by hydroformylation, that is reaction with carbon monoxide and hydrogen, according to the Oxo process. Catalysts employed can be cobalt or rhodium which may be modified with phosphine, phosphite, arsine or pyridine ligands, as described in U.S. Pat. Nos. 3,231,621; 3,239,566; 3,239,569; 3,239,570; 3,239,571; 3,420,898; 3,440,291; 3,448,158; 3,448,157; 3,496,203; and 3,496,204; 3,501,515; and 3,527,818.

[0028] Typical hydroformylation reaction conditions include a temperature of about $125^{\circ}C$ to about $200^{\circ}C$, a pressure of about 2170 kPa to about 32550 kPa (300 psig to 4000 psig) and a catalyst to olefin ratio of about 1:5000 to about 1:1.

The molar ratio of hydrogen to carbon monoxide is usually about 0.5 to about 10, such as about 1 to about 2. The hydroformylation reaction typically produces an aldehyde which can then be hydrogenated to generate the required alcohol product.

[0029] The hydroformylation process can be carried out in the presence of an inert solvent, such as a ketone, e.g., acetone, methyl ethyl ketone, methyl isobutyl ketone, acetophenone, and cyclohexanone; an aromatic compound, e.g., benzene, toluene and the xylenes; a halogenated aromatic compound, e.g., chlorobenzene and orthodichlorobenzene; a halogenated paraffinic hydrocarbon, e.g., methylene chloride and carbon tetrachloride; a paraffin, e.g., hexane,

Suitable aromatic hydrocarbons include benzene, toluene, xylene and naphthalene, with preferred compounds being benzene and toluene.

[0033] In one embodiment, the catalyst is a homogeneous acid catalyst such as a Lewis acid catalyst, for example aluminum chloride. Alternatively, the homogeneous acid catalyst is a Brønsted acid catalyst, such as HF or phosphoric acid. Suitable alkylation conditions with a homogeneous catalyst include a temperature of from about -10°C to about 100°C , a pressure of from about 100 kPa to about 2500 kPa (1.0 to 25 atmospheres), a feed weight hourly space velocity (WHSV) of from about 0.2 hr^{-1} to about 10 hr^{-1} and an aromatic compound to olefinic hydrocarbon mixture mole ratio of from about 1:1 to about 15:1. Typical reaction conditions include a temperature of from about 0°C to about 50°C , a pressure of from about 100 kPa to about 300 kPa (1.0 to about 3.0 atmospheres), a feed weight hourly space velocity (WHSV) of from about 0.1 hr^{-1} to about 0.5 hr^{-1} and an aromatic compound to olefinic hydrocarbon mixture mole ratio of from about 5:1 to about 10:1. The reactants can be in either the vapor phase or the liquid phase and can be neat, i.e., free from intentional admixture or dilution with other material, or they can be brought into contact with the catalyst composition with the aid of carrier gases or diluents such as, for example, hydrogen or nitrogen.

[0034] In a further embodiment, the alkylation process is conducted in the presence of a heterogeneous catalyst, such as a molecular sieve. Suitable molecular sieves include mordenite, particularly dealuminized mordenite and other 6-7 Angstrom pore molecular sieves disclosed in U.S. Patent No. 5,026,933.

[0035] In one practical embodiment, the alkylation catalyst comprises a molecular sieve having an X-ray diffraction pattern including d-spacing maxima at 12.4 ± 0.25 , 6.9 ± 0.15 , 3.57 ± 0.07 and 3.42 ± 0.07 Angstroms. The X-ray diffraction data used to characterize said molecular sieve are obtained by standard techniques using the K-alpha doublet of copper as the incident radiation and a diffractometer equipped with a scintillation counter and associated computer as the collection system. Materials having the required X-ray diffraction lines are sometimes referred to as molecular sieves of the MCM-22 family and include

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MCM-22 (described in U.S. Patent No.4,954,325), PSH-3 (described in U.S. Patent No. 4,439,409), SSZ-25 (described in U.S. Patent No. 4,826,667), ERB-1 is described in European Patent No.0293032, ITQ-1 is described in U.S. Patent No 6,077,498, ITQ-2 is described in International Patent Publication No. WO97/17290, MCM-36 (described in U.S. Patent No. 5,250,277), MCM-49 (described in U.S. Patent No. 5,236,575) and MCM-56 (described in U.S. Patent No. 5,362,697).

[0036] The molecular sieve alkylation catalyst can be combined in conventional manner with an oxide binder, such as alumina, such that the final alkylation catalyst contains between about 2 and about 80 wt% sieve.

[0037] With a molecular sieve catalyst, suitable alkylation conditions include a temperature of from about 0°C to about 500°C, a pressure of from about 20 kPa to about 25000 kPa (0.2 to 250 atmospheres), a feed weight hourly space velocity (WHSV) of from about 0.1 hr⁻¹ to about 500 hr⁻¹, and an aromatic compound to olefinic hydrocarbon mixture mole ratio of from about 1:1 to about 20:1. The WHSV is based upon the weight of the catalyst composition employed, i.e., the total weight of active catalyst (and binder if present). Typical reaction conditions include a temperature within the range of from about 100°C to about 350°C, a pressure of from about 100 kPa to about 2500 kPa (1 to 25 atmospheres), a WHSV of from about 0.5 hr⁻¹ to about 100 hr⁻¹ and an aromatic compound to olefinic hydrocarbon mixture mole ratio of from about 4:1 to about 15:1. Again, the reactants can be in either the vapor phase or the liquid phase and can be neat, i.e., free from intentional admixture or dilution with other material, or they can be brought into contact with the zeolite catalyst composition with the aid of carrier gases or diluents such as, for example, hydrogen or nitrogen.

[0038] The alkylation process of the invention produces an alkylaromatic hydrocarbon mixture in which the alkyl side chains are lightly branched and have less than 0.5 atom% of quaternary carbon atoms and in which most of the aromatic species are located at the 2- or 3- position in the alkyl side chain. The alkylaromatic hydrocarbon mixture is therefore particularly useful as an intermediate in the production of alkylarylsulfonates, which are useful as detergents or surfactants. Processes for sulfonating alkylbenzenes are described

in the U.S. Patent No. 4,298,547. More particularly, alkylaromatic hydrocarbons may be converted to alkylarylsulfonates by sulfonation of the aromatic ring with sulfuric acid. The sulfonation reaction is well known in the art and is commonly carried out by contacting the organic compound with sulfuric acid at temperatures of from about -70°C to about $+60^{\circ}\text{C}$. Detailed descriptions of specific commercial processes abound in the literature. See, for instance, pages 60-62 of INDUSTRIAL CHEMICALS, Third Edition, by W. L. Faith et al, published by John Wiley & Sons, Inc.

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CLAIMS

1. An olefin oligomerization process comprising:
 - (a) contacting a feedstock comprising one or more C_2 to C_6 n-olefins and from about 0.5 wt% to about 25 wt% of an iso-olefin under oligomerization conditions with surface-deactivated ZSM-23 to produce an oligomerized olefin product having a $C_{12}+$ fraction containing less than 0.5 atom % of quaternary carbon atoms; and
 - (b) separating from said oligomerized olefin product such $C_{12}+$ fraction.
2. The process according to claim 1, wherein said iso-olefin is iso-butylene and/or iso-amylene.
3. The process according to any preceding claim, wherein said one or more n-olefins in the feedstock are selected from propylene, n-butene and mixtures thereof.
4. The process according to any preceding claim, wherein said feedstock is the unreacted effluent stream from an MTBE unit.
5. The process according to any preceding claim, wherein said feedstock contains less than 100 ppm of dimethyl ether.
6. The process according to any preceding claim, wherein said feedstock has a sulfur content of less than 10 ppm.
7. The process according to any preceding claim, wherein said ZSM-23 has been surface deactivated with a sterically hindered nitrogenous base.

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8. The process according to Claim 7, wherein said sterically hindered nitrogenous base is 2,4,6-collidine.
9. The process according to any preceding claim, wherein said oligomerization conditions include a temperature of 160 to 250°C.
10. The process according to any preceding claim, wherein said oligomerization conditions include a temperature of 190 to 230°C.
11. The process according to any preceding claim, wherein said oligomerization conditions include a temperature of 210 to 220°C.
12. The process according to any preceding claim, wherein said oligomerization conditions comprise a pressure in the range of from 500 psig (3447 kPa (gauge)) to 1500 psig (10342 kPa (gauge)).
13. The process according to any preceding claim, wherein said oligomerization conditions comprise a pressure in the range of from 750 psig (5171 kPa (gauge)) to 1250 psig (8618 kPa (gauge)).
14. The process according to any preceding claim, wherein said oligomerization conditions comprise a weight hourly space velocity of from 0.1 hr⁻¹ to 4.0 hr⁻¹.
15. The process according to any preceding claim, wherein said oligomerization conditions comprise a weight hourly space velocity of from 0.2 hr⁻¹ to 3.0 hr⁻¹.
16. The process according to any preceding claim, wherein said oligomerization conditions comprise a weight hourly space velocity of from 1.75 hr⁻¹ to 2.25 hr⁻¹.

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17. The process according to any preceding claim, wherein said $C_{12}+$ fraction has an average of from 0.8 to 2.0 C_1 - C_3 alkyl branches per carbon chain.
18. The process according to any preceding claim, wherein said $C_{12}+$ fraction has an average of from 0.8 to 1.3 C_1 - C_3 alkyl branches per carbon chain.
19. A method for producing a long chain alcohol mixture comprising contacting the $C_{12}+$ fraction produced by the process of any preceding claim with carbon monoxide and hydrogen under hydroformylation conditions and in the presence of a hydroformylation catalyst.
20. A method for producing an alkylaromatic compound comprising contacting an aromatic compound with the $C_{12}+$ fraction produced by the process of any one of claims 1 to 18 under alkylation conditions and in the presence of an alkylation catalyst.
21. A method for preparing an alkylaryl sulfonate by sulfonating the alkylaromatic compound produced by the method of Claim 20.

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